## Frequency comb spectroscopy on trapped calcium ions

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# Summary

Frequency is currently the physical quantity which can be measured to the highest precision. The record in accuracy for such a measurement was obtained in a comparison of the $^1S_0 - ^3P_0$ transition in two $^{27}Al^+$ clocks, where an inaccuracy of 9 parts in $10^{18}$ was obtained [1]. The described experiment uses the combination of a Paul trap for confinement and a frequency comb for calibration. This combination provides an ideal tool for spectroscopy experiments, because the ions in a trap can be directly or sympathetically cooled and studied with a long interaction time, while a frequency comb adds a broad range of wavelengths for spectroscopy. In the work described in this thesis, we combine the advantages of ion trapping with (direct) frequency comb spectroscopy. We trap and study $^{40}Ca^+$, which is of interest for applications in ion clocks, the search for a variation of the fine-structure constant and quantum computing, as is described in Chapter 1.

For our experiments, two types of linear Paul traps have been constructed, which are discussed in Chapter 2. A linear Paul trap uses electric fields to trap ions. The field is formed by a combination of oscillating Radio Frequency (RF) fields for radial confinement, and DC electric fields on so-called endcaps to trap the ion in the axial direction. The first ion trap that we have constructed uses four elongated RF electrodes for radial trapping, and two ring electrodes to which the endcap potential is applied. Ions are made from a neutral beam of evaporated calcium atoms, which is photo-ionized inside the trapping region. The trapped ions are laser cooled using only diode lasers, with the combination of a cooling laser at 397 nm on the $4s\,^2S_{1/2} - 4p\,^2P_{1/2}$ transition and a repumper laser at 866 nm on the $3d\,^2D_{3/2} - 4p\,^2P_{1/2}$-transition. With this method calcium ions have succesfully been trapped and laser cooled to crystallization. The first trap has been employed in two experiments on dipole-allowed transitions in calcium ions, which are described in Chapter 5 and 6.

A second ion trap has been developed with a combination of two RF electrodes that extend over the full length of the trap, and two segmented electrodes which serve both as endcap electrodes and for correction of patch potentials. In this trap calcium ions are made and laser cooled using the same methods as described for the first trap. The second trap has been employed for an experiment on direct frequency comb spectroscopy of a dipole-forbidden (clock) transition in $^{40}Ca^+$, as is described in Chapter 7.

Besides the ion traps, two frequency combs have been constructed, for the purpose of high precision spectroscopy of transitions in the trapped ions. These combs are the subject of chapter 3. Both frequency combs are based on a Ti:Sa laser, in which Kerr-lens mode locking together with group velocity dispersion control and a broadband gain spectrum makes the generation of ultrashort pulses possible. We have built a laser with a broadband spectrum that is used as a seed laser for a noncollinear optical parametric chirped pulse amplifier, and a laser with a narrower spectrum for the purpose of direct frequency comb spectroscopy.

A frequency comb can be used in two ways to calibrate a transition frequency: either through an intermediate cw laser, or by directly exciting the transition of interest. In the first case, a resonance is detected using the cw laser, while the laser frequency is calibrated against the frequency comb. In the case of direct frequency comb spectroscopy, the comb is used both for excitation and calibration. However, the power from a comb mode usually is orders of magnitude lower than for the case with a cw laser. For a one-photon transition, an additional disadvantage is the presence of a large number of background modes, while only one mode at a time can excite the transition. We have performed both types of experiments on transitions in calcium ions, using the method of shelving to overcome the signal-to-noise issues of direct frequency comb spectroscopy.

The three frequency metrology experiments that we performed on trapped and cooled calcium ions are described in Chapter 5-7. The first experiment is the calibration of the $4s\,^2\mathrm{S}_{1/2} - 4p\,^2\mathrm{P}_{1/2}$ transition, which is described in Chapter 5. In this experiment, a frequency doubled diode laser is used to excite the transition, while the fundamental laser wavelength at 794 nm is calibrated against a frequency comb. The ion fluorescence is measured using a Photomultiplier Tube (PMT). The total measurement, accounting for the measured systematic effects, yielded a transition frequency of $\nu = 755\,222\,766.2\,(1.7)$ MHz. The obtained accuracy presents an order of magnitude improvement over the previous most accurate result.

A second spectroscopy experiment was conducted on the $4s\,^2\mathrm{S}_{1/2} - 4p\,^2\mathrm{P}_{3/2}$ transition at 393.5 nm, which is the subject of Chapter 6. For the excitation of the transition a frequency comb was employed directly, which was frequency doubled at full repetition rate. The signal-to-noise issue described earlier is overcome by employing a shelving scheme, using the 7 % chance of excited ions to decay to the long-lived $3d\,^2\mathrm{D}_{5/2}$ energy level. By monitoring the cooling laser fluorescence with a PMT, while the $4s\,^2\mathrm{S}_{1/2} - 4p\,^2\mathrm{P}_{3/2}$ transition is resonantly excited with the frequency comb, the excitation can be indirectly detected through a decrease of fluorescence on the cooling transition. The cooling laser fluorescence is alternately measured with and without a repumper on the $3d\,^2\mathrm{D}_{5/2} - 4p\,^2\mathrm{P}_{3/2}$ transition,

collecting both signal and background. These two datasets are subtracted and corrected for loss of ions from the trap. The determined $4s\,^2\mathrm{S}_{1/2} - 4p\,^2\mathrm{P}_{3/2}$ transition frequency in $^{40}\mathrm{Ca}^+$ is $\nu = 761\,905\,012.7\,(0.5)$ MHz, improving the best previously known value by more than two orders of magnitude.

In Chapter 7 a third experiment is described, where direct frequency comb spectroscopy applied to a dipole-forbidden clock transition: the $4s\,^2\mathrm{S}_{1/2} - 3d\,^2\mathrm{D}_{5/2}$ transition at 729.5 nm. The natural linewidth of this transition is 0.14 Hz which, combined with the low power per comb mode in modes of $\sim$ 2 MHz linewidth, leads to a very low excitation rate of $\sim$ 7 per ion per 1000 seconds. Despite this low rate, the transition can still be detected by again using shelving in the $3d\,^2\mathrm{D}_{5/2}$ state. A string of 10 calcium ions is created, and its cooling laser fluorescence is imaged onto an electron-multiplying CCD camera. When an ion is excited to the $3d\,^2\mathrm{D}_{5/2}$ state, it will turn dark in the camera image for the lifetime of the ion of about 1 second, since it no longer participates in the cooling cycle. The dark ions are detected, and the rate of ions turning dark is measured as a function of the excitation wavelength. Using this method, we found a transition frequency of $\nu = 411\,042\,129.6\,(0.3)$ MHz, which is consistent with previously reported measurements [27]. The accuracy and measurement rate of this experiment is currently limited by the Doppler temperature of the trapped ions, in combination with the linewidth of the comb modes. It can be improved upon by trapping and probing the ions in the Lamb-Dicke regime, in combination with a reduction of the comb mode linewidth by locking to a high-finesse optical cavity.

In conclusion, in this thesis the versatility of (direct) frequency comb spectroscopy in an ion trap is demonstrated through several experiments on calcium ions. We have shown that transitions can be calibrated both in the infrared and in the visible frequency regime. These experiments demonstrate the wide range of wavelengths that can be accessed for direct frequency comb spectroscopy using a single laser. In addition, the method of direct frequency comb spectroscopy has been applied to measure both a dipole allowed and an electric quadrupole transition. These results illustrate the possibility to employ the combination of a frequency comb and an ion trap for a wide range of calibration purposes.